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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Technical Note: Mineralogical, chemical, morphological, and optical interrelationships of mineral dust re-suspensions" *by* Johann P. Engelbrecht et al.

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The technical note presents results from measurements of surface soils collected from locations all over the world, which are representative for many dust source regions. Various techniques were applied to analyze the mineral and chemical composition as well as the morphological and optical properties of the soil samples. The size distributions of the soils were determined after treatment (sieving and suspending in an aqueous solution). The data provided in this study allow insight into the variability of the properties of dust generating soils in different source regions, and they are potentially very useful for remote sensing and dust modeling purposes. I have only a few minor

remarks, which should be taken into consideration for the publication of the manuscript.

In Section 1, on page 1–2, the authors state that only particles smaller than 10 μm generally survive long-range transport. Although this may be true for very large distances, some studies found a significant volume fraction of dust with particles sizes larger than 10 μm after transport (Stuut et al., 2005; Jeong et al., 2014). Thus, it may be better to not phrase this as absolutely and to also mention these other studies.

Authors' Response: We agree. This was also pointed out by Reviewer 1. The sentence was rephrased and additional references (Stuut et al 2005, Jeong et al., 2014) added (see Reviewer 1 first comment) to demonstrate that larger particles have been known to be transported over longer distances. Authors' changes (additions) to manuscript In the past only particles less than $\approx$ 10 μm in diameter were considered to survive long-range transport in the atmosphere (Maring et al., 2003;Zender et al., 2003;Formenti et al., 2011). However, multiple studies have shown that larger mineral dust particles can be transported over distances of hundreds to thousands of kilometres (Alastuey et al., 2005;Betzer et al., 1988;Lawrence and Neff, 2009;McTainsh et al., 1997;Neff et al., 2013;Menéndez et al., 2014). Quartz particles, up to $\approx$ 160 μm in diameter, ascribed to Saharan dust storms, were reported from deposits on La Graciosa Island (Menéndez et al., 2014), about 160 km off the west coast of Africa. Samples of airborne dust collected off the west African coast (Stuut et al., 2005) had mean diameters 8-42 μm, with particles up to 200 μm in diameter being identified. Asian dust transported to Korea over a distance of 2000 km contained a substantial proportion of giant mineral particles up to 60 μm in diameter, of clay aggregates as well as clay coated quartz, feldspar, and mica grains (Jeong et al., 2014). Complex aggregates (iberulites) of up to 100 μm in diameter, were suggested to have formed in the atmosphere during long range transport of mineral dust from Saharan and Sahel, to southern Spain (Díaz-Hernández, 2008). Such aggregates were found to be composed largely of quartz, feldspars, and carbonates, in a matrix of various clay minerals (Cuadros et al., 2015).

In Section 2.1, on page 2, it is mentioned that the soil samples were first sieved to

remove larger pebbles, and then additionally sieved to obtain sub-sets of different size classes of the soils. Soil consists of aggregates to a large degree, and so does emitted airborne dust, despite partial fragmentation of the aggregates that occurs during dust emission (Kok , 2011). The literature states that some sieving techniques, especially wet sieving disperse the aggregates more than others, shifting the sieved soil particle size distribution to smaller sizes, compared to the non-dispersed parent soils in the dust source regions (Shao, 2001). How much dispersion occurs due to wet sieving also depends on the soil type (Choate et al., 2006). Thus, the dispersion of aggregates by sieving techniques that are commonly applied for soil analyses may introduce a bias into the analysis with respect to the size distribution of specific dust/mineral properties, including the ones measured after re-suspension of the soil. For instance, the abundance of clay minerals in the smaller size ranges may be increased at the expense of the larger size ranges in the measurements, compared to the undisturbed distribution in soils or airborne dust. The authors should add information on what sieving techniques were applied by them. Also, it should be discussed how the techniques may have affected the measurements due to possible dispersion of the aggregated particles. Such information will be helpful for interpretation, for instance when the data are used for modeling studies.

Authors' Response: We agree that the sieving, especially wet sieving will de-segregate the soil aggregates and overestimate the fine fractions. Since we only dry sieved all our samples, we consider it unnecessary to discuss the disadvantages of wet sieving. However, we will clarify this with the following additions to the text. Authors' changes (additions) to manuscript All samples were dry-sieved to retain their mineralogical and physical integrity, as close as possible to the surface soils from which they were collected. Wet-sieving was avoided since that would dissolve evaporite minerals, including most chlorides and sulfates contained in many soils, and also disaggregate clay clusters into smaller fragments, thereby creating a bias towards the finer particle sizes (Choate et al., 2006). The re-suspension of dry-sieved samples in the dust entrainment facility, better resembles the generation of dust from exposed natural soils by particle

bombardment, attrition, and fragmentation (Kok, 2011;Shao, 2001).

In Section 3.2.2, page 11, the study by Lafon et al. (2006), where estimates for the Fe/Al-ratio in dust are provided for Chinese, North African, and Sahelian dust, should be referenced and included in the comparison.

Authors' Response: Thanks for pointing this out, we now include this study in our discussion. We added the following at the top of page 12.

Authors' changes (additions) to manuscript Dust samples from Niger, Cape Verde, Tunisia, and China, suspended on polycarbonate filters, before being analyzed by XRF, gave Fe2O3 concentrations of 6.2 – 8.7 %, and Fe/Al ratios of 0.43 – 6.0 (Lafon et al., 2006). We suggest that differences in aluminum (Al) and iron (Fe) concentrations, and subsequent range in Fe/Al ratios found by investigators, not only to be due to chemical and mineralogical differences amongst the samples, but can in part be ascribed to differences in sampling and analytical procedures.

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R. Reynolds (Referee 2) rreynolds@usgs.gov Received and published: 21 June 2016 General comments This Technical Note provides a compendium of important mineralogic, chemical, physical properties of widely distributed materials (mostly surface soils and aeolian sediments) sampled from re-suspension in the PM2.5 and PM10 size fractions. The primary goal is to provide data that can be used to evaluate the effects of atmospheric dust primarily with respect to radiative forcing and human health.

The measurement of radiative properties during re-suspension and comparison with compositional parameters provides important information relating dust mineralogy to optical properties (Fig. 4.5-1). The SEM micrographs and accompanying EDS spectra are of exceptional quality and provide a helpful view into typical mineralogic complexity of PM2.5 particles. The comparisons among SSAs, iron, and clay minerals invite collaborative discussions about alternative explanations for the observed relations. Specific comments. The manuscript emphasizes the relations among iron, clay minerals, and optical properties. Fe is interpreted to mostly reside in montmorillonite on the basis of correlation of Fe and Al (p. 11). Certainly, some Fe is structurally bound in many of the clay minerals encountered in this study. But how much? A quick scan of literature and text books indicate typical structural Fe concentrations in montmorillonite and illite between _1 and 7 wt %, expressed as Fe2O3. (Of course, chlorite and nontronite by classification and structure contain a lot of Fe – on the order of 8-20+ wt % Fe2O3.) Considering the samples that contain primarily rock-derived sediment (i.e., ignoring salt-/carbonate- /diatom-bearing playa samples), the correspondence between Al and Fe could also be attributed to the co-existence of ferric oxide minerals (hematite and goethite) with clay minerals, in varying relative proportions depending on factors of provenance, sorting, and pedogenesis, in some instances. One reason for my concern (with background in iron oxides but not in clay minerals) stems from the relations illustrated in the scatter plots of Fig. S4-4. The ratios of Fe2O3 to Al2O3 for most samples (blue markers) seem to be about 0.7. Some sleuthing on chemical analyses of clays, including "standards" used for reflectance spectroscopy (http://speclab.cr.usgs.gov/spectral.lib06/ds231/datatable.html) reveals the following Fe2O3/Al2O3 ratios: illite: 0.07-0.29 montmorillonite: 0.04-0.28 kaolinite: 0.01-0.04 chlorite: 0.48 These ratios are based on the following analyses for Fe2O3 wt %: Illites: 1.6, 6.4, 4.99 Montmorillonite: 5.8, 3.2, 3.5, 1.4, 1.75, 1.2, 3.85 Chlorite 8.3 In many of this study's samples, there seems to be a large excess of chemical Fe2O3 that cannot represent structural Fe in clays. I suggest that the authors reconsider their interpretation that the correspondence between Fe and Al is produced primarily by

structural Fe in clay minerals. I suggest instead that – except perhaps for the samples low in Fe and Al – the "excess Fe" is better explained by the presence of the minerals hematite and goethite these sediments, beyond their abundance attributed by the authors. The reason that these ferric oxides have not been commonly noted is that researchers have not looked closely enough for them with specialized instrumentation, which is admittedly difficult to access. These minerals appear to be common in fine-grained, rock-derived sediments worldwide. They are probably ubiquitous in sediments having hues of red, orange, and yellow. Recent work shows these ferric oxides to be commonly nano-sized (<50 nm) and to occur as discrete particles within and on clay minerals and in other occurrences in the same sample. They are revealed by certain magnetic properties and Mössbauer spectroscopy especially at 4.2K (liquid He temperature) because these ultrafine minerals are magnetically ordered at very low temperatures but not at room temperature. The nano-ferric-oxide minerals can be observed (and confirmed as Fe oxide) under very high resolution SEM with EDA beam diameters on the order of a few 10s nm. I suggest that the dust community consider the following: The radiative properties of at least many samples (standards and natural) described as containing Fe-bearing clays are actually strongly controlled by the presence of ferric oxide minerals, many of which are nano-size (Reynolds et al., 2014a, 2014b; Moskowitz, manuscript in review). This distinction is no small matter with respect to modeling the radiative effects of dust in the atmosphere and on surfaces of ice and snow. The authors similarly attribute the correspondences between Ti - Al and Mn – Ti to residence of Ti and Mn in clays. It appears to me that much of the Ti and Mn are associated with Fe(-Ti) oxide minerals such as ilmenite (usually <5 wt %) and (or) the generally more abundant ferromagnesium silicates, as identified in many samples.

Response: This is an important observation and we greatly appreciate the reviewer's concerted effort at not only pointing this out but also providing supporting literature. We added a table (Table 2) to the manuscript to illustrate differences between our chemical results and published data on clay minerals. To better describe the discrepancy of the Fe and other elements in the dust re-suspensions, the following is added at the end of

paragraph 3.2.2, after line 2 on page 12. Authors' changes (additions) to manuscript The high correlations between iron (Fe) and aluminum (Al) are attributed to the close co-existence of oxihydroxide minerals (hematite, goethite, magnetite (Fe3O4)) with clays and clay-like phases, micas, chlorite, and other silicates, all in varying proportions, depending on factors of provenance, chemical weathering, and pedogenesis. Comparisons of chemical results of re-suspended soils from this study, and those published on biotites (Deer et al., 1962) and clays (Weaver and Pollard, 1973) are shown in Table 2. Clay minerals identified in this study, by XRD and SEM, include illite, palygorskite and kaolinite, with montmorillonite in a few samples. The average aluminum (Al) concentrations of our re-suspended soils (3.58% for PM10, 4.64% for PM2.5) (Table 2(a), 2(b)) are substantially lower than those of chlorite (22.06%) (Table 2(d)), illite (13.94%) (Table 2(e), montmorillonite (11.6%) (Table 2(f) or palygorskite (6.73%) (Table 2(g)). On the other hand, the iron (Fe) contents of the re-suspended soils (ave. 3.33% for PM10, 3.65% for PM2.5) are less or similar to those of chlorite (2.21%), illite (3.47%), montmorillonite (2.79%), or palygorskite (2.26%). The resultant Fe/Al ratios for the re-suspensions are proportionally greater (ave. 1.01 for PM10, 0.87 for PM2.5) than those of chlorite (0.07), illite (0.25), montmorillonite (0.24) or palygorskite (0.27) (Table 2 (d)-(g)). The higher Fe/Al ratios for the re-suspended soil samples point to the presence of Fe-bearing minerals other than clays in the particles. It is evident that although some iron (Fe) is contained in the crystal structures of the chlorite, illite, montmorillonite, palygorskite, and other clay minerals, the remaining iron (Fe) is contained in oxihydroxides such as hematite (Fe2O3), goethite (FeO.OH), or magnetite (Fe3O4), as well as iron bearing silicates such as biotite (Table 2(b), amphibole, and pyroxene, all minerals identified in the sieved soil fractions by optical microscopy and XRD. Also, the grouping of the majority (122) of the samples (Figure 17) and the high correlation coefficient of 0.79 between iron (Fe) and aluminum (Al) (Table 1), point to a strong mineralogical interrelationship amongst the Fe-bearing clays, Fe-oxihydroxides (goethite, hematite, magnetite), and silicates such as micas (biotite, muscovite) and chlorite. Recent studies (Reynolds et al., 2014a;Reynolds et al., 2014b;Kars et al., 2015), applying

a combination of Mössbauer spectroscopy, magnetic measurements, reflectance spectroscopy, and scanning electron microscopy (SEM), provide evidence of nanometer size (< 20 nm) particles of hematite, goethite, and magnetite, closely associated with clay-like particles. Similarly, from magnetic measurements on the Callovian-Oxfordian claystones in the Paris Basin, Kars et al. (2015) suggest nano-particles of goethite to be dispersed in a clay-like matrix. Fordham (1990), from electron microscopy and XRD investigations concluded that biotite underwent chemical weathering at particle edges and along cleavage plains, to form finely divided interstratified flakes of illite and iron oxihydroxides set in a clay-like matrix. Similarly, Borchard et al. (1971) had identified colloidal coatings of amorphous clay-like particles on volcanic ash in soils. To understand the elevated concentrations of iron (Fe), titanium (Ti) calcium (Ca), and potassium (K), as well as Fe/Al ratios in our soil re-suspensions, compared to those in crystalline clays (illite, montmorillonite and palygorskite) (Table 2), we propose that particles of PM10 and PM2.5 are largely composed of nano-size particles of micas, clays, and metal oxides, to be dispersed as a colloidal particles, and the ions to be adsorbed in an amorphous clay-like matrix. We suggest these colloidal composites to be residual products of biotite and other ferro-magnesian particles, formed by chemical weathering and pedogenesis, forming particle coatings, individual particles, aggregates, and interstitial material, with quartz, feldspars, and other silicates.

Table 2. Average concentrations and elemental ratios for (a) PM10 and (b) PM2.5 re-suspensions from this study, (c) biotite, (Deer et al., 1962, Table 13), chlorite (Weaver and Pollard, 1973, Table XLII), Illite (ditto, Table III), montmorillonite (ditto,Table XXV), and palygorskite (ditto, Table LII).

Issues of "long-range transport" [not defined] of dust particles "typically < 9 microns": A body of work indicates the presence of larger particles transported at regional (10s to many 100s km) and global scales (many 1000s km). A few articles on the topics are: Beltzer et al., 1988; McTainsh et al. 1997; Middleton et al., 2001; Alastuey et al 2005, Chan et al., 2005; Lawrence and Neff, 2009; Neff et al., 2013; van der Does

et al., 2015. Particles >PM10 certainly have important effects on the Earth System through radiative properties (atmospheric and snow/ice melt), infrastructure, health (in the upper respiratory system if not deeper), and fertilization. Was there any black carbon in any of the samples that might have enhanced measured radiative properties? P. 2, lines 8-9. Authors' Response This was also pointed out by other reviewers. The following was added.

Authors' changes (additions) to manuscript In the past only particles less than $\approx$ 10 μm in diameter were considered to survive long-range transport in the atmosphere (Maring et al., 2003;Zender et al., 2003;Formenti et al., 2011). However, multiple studies have shown that larger mineral dust particles can be transported over distances of hundreds to thousands of kilometres (Alastuey et al., 2005;Betzer et al., 1988;Lawrence and Neff, 2009;McTainsh et al., 1997;Neff et al., 2013;Menéndez et al., 2014). Quartz particles, up to $\approx$ 160 μm in diameter, ascribed to Saharan dust storms, were reported from deposits on La Graciosa Island (Menéndez et al., 2014), about 160 km off the west coast of Africa. Samples of airborne dust collected off the west African coast (Stuut et al., 2005) had mean diameters 8-42 μm, with particles up to 200 μm in diameter being identified. Asian dust transported to Korea over a distance of 2000 km contained a substantial proportion of giant mineral particles up to 60 μm in diameter, of clay aggregates as well as clay coated quartz, feldspar, and mica grains (Jeong et al., 2014). Complex aggregates (iberulites) of up to 100 μm in diameter, were suggested to have formed in the atmosphere during long range transport of mineral dust from Saharan and Sahel, to southern Spain (Díaz-Hernández, 2008). Such aggregates were found to be composed largely of quartz, feldspars, and carbonates, in a matrix of various clay minerals (Cuadros et al., 2015).

Please see Ghio et al., 2014 with respect to medical studies of biologic effects of dust on human lung cells.

Response: Reference was added

One comment about presentation that I did not include in my previous "comments": I think that the section (3.1) on samples and settings could be organized more tightly among the sub-sections (e.g., 3.1.1, 3.1.2, 3.1.3,....3.1.25). Some samples are described in adequate detail but others are very vague (e.g., "The two samples from Utah were similar to each other, both taken along a dirt road" does not convey any meaningful information and leaves open many questions–what are the local substrates; was the road graveled and (or) graded? And more. With parallelism and more consistency, Section 3.1 would be more easily read and informative. Response: We added more text to some of these site descriptions, as suggested. We had grouped the samples by distinctive soil type, e.g. diatomaceous silts, or by geographical terrain. It will be difficult to group them further, and therefore we retained the original sub-sections. 3.1.7 Southwestern U.S.A. – Ft. Carson, CO (S3011) Authors' changes (additions) to manuscript Fort Carson is located in central Colorado at the foot of the Rocky Mountains and to the south of Colorado Springs. The area is largely underlain by fluvial and alluvial clays, silts, and sands. The surface soil sample was collected from an unpaved road on the military base. It contains major amounts of quartz and plagioclase, with lesser amounts of biotite, potassium feldspars, calcite, hornblende, muscovite, dolomite, gypsum, and clays (Engelbrecht et al., 2012).

3.1.8 Southwestern U.S.A. – Dugway, UT (S3016, S3017) Authors' changes (additions) to manuscript The two samples from Utah are similar to each other, both collected along built-up dirt roads on the military base, considered to be local dust sources of concern. The road gravel had been excavated from a local quarry. Both samples contain major amounts of quartz and calcite (Figure 5), and lesser amounts of dolomite, aragonite, plagioclase, with traces of biotite, muscovite, illite, and orthoclase.

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Dear Sir, As expected, re-suspended mineral dust aggregates studied by the authors have the general features of soil particles (Figs. 4-10 and 12-14). According to these authors, only particles less than _ 10 μm in diameter survive long-range transport; however, the giant particles reported by Jeong et al. (2014) were transported directly from source. At the contrary, mineral aggregation is not limited to soil-forming processes, because the aggregation of fine mineral particles observed in some airborne particles may occur via atmospheric processes. Iberulites result from aggregation phenomena in atmosphere when Saharan dust outbreaks reach the South of Spain (Diaz-Hernandez and Parraga, 2008). However, some textural, mineralogical and compositional features observed for these giant airborne dust particles are similar to those of the re-suspended soils particles described by the authors (see Cuadros et al., 2015). We propose that these considerations should be included in the Introduction and Conclusions in relation to mineral aggregates observed in dust particles

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Authors' Response We agree with the reviewer's comments and elaborated further on our statement in the Introduction, page 1 Authors' changes (additions) to manuscript In the past only particles less than $\approx$ 10 μm in diameter were considered to survive long-range transport in the atmosphere (Maring et al., 2003;Zender et al., 2003;Formenti et al., 2011). However, multiple studies have shown that larger mineral dust particles can be transported over distances of hundreds to thousands of kilometres (Alastuey et al., 2005;Betzer et al., 1988;Lawrence and Neff, 2009;McTainsh et al., 1997;Neff et al., 2013;Menéndez et al., 2014). Quartz particles, up to $\approx$ 160 μm in diameter, ascribed to Saharan dust storms, were reported from deposits on La Graciosa Island (Menéndez et al., 2014), about 160 km off the west coast of Africa. Samples of airborne dust collected off the west African coast (Stuut et al., 2005) had mean diameters 8-42 μm, with particles up to 200 μm in diameter being identified. Asian dust transported to Korea over a distance of 2000 km contained a substantial proportion of giant mineral particles up to 60 μm in diameter, of clay aggregates as well as clay coated quartz, feldspar, and mica grains (Jeong et al., 2014). Complex aggregates (iberulites) of up to 100 μm in diameter, were suggested to have formed in the atmosphere during long range transport of mineral dust from Saharan and Sahel, to southern Spain (Díaz-Hernández, 2008). Such aggregates were found to be composed largely of quartz, feldspars, and carbonates, in a matrix of various clay minerals (Cuadros et al., 2015).

Also added in the Conclusion, page 13, line 10

However, it is recognized that dusts may be modified as they mix with other aerosols, and age under changing meteorological conditions and over time, in some cases forming larger aggregates of particles. In our study, SEM-based secondary electron images of the re-suspended dust samples are evidence of such mineral aggregates, as well as of coatings on quartz, feldspar, and other mineral grains, similar to those reported for ambient aerosols (Cuadros et al., 2015;Díaz-Hernández, 2008;Engelbrecht et al., 2009;Jeong et al., 2014).

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